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Interim Guidelines for PAH Contamination at Abandoned Coal Tar Sites

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INTERIM GUIDELINES

FOR PAH CONTAMINATION

AT ABANDONED COAL TAR SITES

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PREPARED FOR THE

Waste Management Committee
Toxic Substances Advisory Committee
of the
Canadian Council of Resource and Environment Ministers

BY THE

Ad Hoc Federal-Provincial Working Group
on Interim PAH Guidelines

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INTERIM GUIDELINES FOR PAH CONTAMINATION AT ABANDONED COAL TAR SITES EXECUTIVE SUMMARY

BACKGROUND

- At their June 1989 meeting, the Toxic Substances Advisory Committee (TSAC) of the Canadian Council of Resource and Environment Ministers (CCREM) directed the Waste Management Committee to strike a working group to develop interim guidelines for polycyclic aromatic hydrocarbons (PAH) associated with coal tar waste sites. A detailed report which outlines the recommended guidelines and rationale has been prepared.

- The development of interim PAH guidelines was undertaken recognizing that several longer term initiatives such as the development of multi-media guidelines for PAH are underway. The interim PAH guidelines developed in this report are, therefore, intended for use by the member governments of the CCREM until definitive criteria are developed.

CONTAMINANTS OF CONCERN

- Coal tar waste sites are contaminated with a complex mixture of toxic chemicals (Appendix A) of which PAH are generally considered to be the most toxic constituent group. The PAH group in turn comprises a number of individual compounds which have a wide range of toxicity. The carcinogenic potential of PAH is the main human health concern.

- For the purpose of developing interim guidelines, the PAH can be divided into two general groups. The substances in Group 1 consist of PAH designated as carcinogenic by the International Agency for Research on Cancer (IARC). The substances in Group 2, although not documented as being carcinogenic are good indicators of coal tar contamination.

- Two additional groups of contaminants (Group 3 and Group 4) are identified as substances of concern at coal tar waste sites. Guidelines for these contaminants have not been developed.

INTERIM GUIDELINES FOR SOIL AND GROUNDWATER

- The principal media subject to contamination by coal tar wastes are soils in the immediate vicinity of the coal tar sites, and the groundwater. Sediment and surface water may also be of concern when the abandoned plant sites are located adjacent to water bodies. Air contamination is of lesser general concern due to the relatively low volitility of the carcinogenic PAH. No guidelines have been recommended by the Working Group for PAH in air, sediments or surface water since existing standards for these media are very limited and not applicable to specific problems associated with contaminated coal tar sites.

- For soil and groundwater, the working group recommends adoption of modified "ABC criteria (Table 1) from the Quebec Ministry of the Environment. The "ABC" criteria for groundwater have been modified to make them more consistent with the World Health Organization (WHO) drinking water guideline of .01 ug/L B(a)P. This value was adopted as being representative of all six carcinogenic PAH and was selected as an "A" value.

- It should be recognized that both the Quebec numbers and the recommended modified ABC values are based on limited toxicological information. The values are based on knowledge of different properties such as toxicity, bio-accumulation potential, etc. and not on risk assessment models.

RECOMMENDED APPLICATION OF GUIDELINES

- The "ABC" approach provides a means to judge whether a site should be considered for further investigation and/or remediation. Depending on the use of the land, different investigative and/or remedial criteria should be considered.

- Investigative criteria are values above which detailed investigation is required to assess the extent of contamination and nature of any hazards, to determine if any remedial actions should be undertaken.

- Remedial criteria are values above which action is required to reduce exposure for humans or other biota. Action could include cleanup, other mitigation and/or change in land use.

Residential or Farming Use

- Investigation Criteria is Value A
- Remedial Criteria is Value B

Commercial or Industrial Use

- Investigative Criteria is the Value B
- Remedial Criteria is Value C

TABLE 1: "ABC" VALUES FOR PAH IN SOIL AND GROUNDWATER AT COAL TAR WASTE SITES

PAH	Conc. in soil (mg/kg dry weight)			Conc. in Groundwater (ug/L)		
	A	B	C	A	B	C
Group 1 Carcinogenic PAH						
benzo(a)anthracene	0.1	1	10	0.01	0.1	1
benzo(b)fluoranthene	0.1	1	10	0.01	0.1	1
benzo(k)fluoranthene	0.1	1	10	0.01	0.1	1
benzo(a)pyrene	0.1	1	10	0.01	0.1	1
dibenz(a,h)anthracene	0.1	1	10	0.01	0.1	1
indeno(1,2,3-cd)pyrene	0.1	1	10	0.01	0.1	1
Group 2 Other PAH						
naphthalene	0.1	5	50	0.2	2	20
phenanthrene	0.1	5	50	0.2	2	20
pyrene	0.1	10	100	0.2	2	20

Value A: This value represents the approximate achievable detection limits for PAH in soil.
In groundwater, value A is based on drinking water criteria as described in the bold-faced paragraph on page 36.

Value A-B: The soil or groundwater is slightly contaminated. At this level of contamination, groundwater generally falls within the range of quality standards and criteria where they exist. However, it is worthwhile to investigate possible sources of contamination, especially in the case of groundwater to ascertain whether new contaminants continue to enter the water. This may lead to intervention focusing on the soil, particularly if the water is used for drinking.
Usually at this level of soil or groundwater contamination, cleanup will not be necessary. However, should the land be redeveloped for especially sensitive purposes, e.g. residential or farming, it may be necessary to implement certain measures, such as the excavation of surficial layers of soil and/or the addition of a layer of clean oil.

Value B: This value is an intermediate value, approximately 5 to 10 times above value A.

Range B-C: The soil or groundwater is contaminated. At this level contamination of groundwater clearly exceeds drinking water standards where they exist and can no longer be used for that purpose.
Although the soil is contaminated, it will not automatically be cleaned up, unless the effect of contaminants on the groundwater necessitates such work.
However, restrictions on land use may be imposed when this level of contamination is observed in the soil. Restoration work may be necessary before the land is used for farming, residential or recreational purposes. Other less sensitive uses, e.g. industrial, commercial and so forth, may be contemplated without clean up being carried out. In all cases, the extent of the work required, e.g. the depth to which soil must be excavated and so on, will depend upon the nature of the contamination, ultimate land use and the impact on groundwater and the environment in general.

Value C: This value is considered to be the level at which contamination is significant.

Range Above C: The soil or groundwater is contaminated. Groundwater cannot be used for drinking. Unless the water is decontaminated, it will have to be monitored closely. Where the soil is contaminated, all uses of such land will be restricted. A thorough analysis must be conducted; in all likelihood, restoration will have to be undertaken before re-development occurs.

APPENDIX A
LIST OF CONTAMINANTS

GROUP 1:	Carcinogenic PAH
	benzo(a)anthracene
	benzo(b)fluoranthene
	benzo(k)fluoranthene
	benzo(a)pyrene
	dibenz(a,h)anthracene
	indeno(1,2,3-cd)pyrene
GROUP 2:	Other PAH
	naphthalene
	phenanthrene
	pyrene
GROUP 3:	Other Organic Compounds
	benzene
	toluene
	xylene
GROUP 4:	Inorganic Compounds
	iron
	arsenic
	sulphide/sulphate
	iron-cyanide complexes
	free cyanide

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Thank you all.

PROPOSED INTERIM GUIDELINES FOR PAH CONTAMINATION AT ABANDONED COAL TAR SITES

1.0 INTRODUCTION

At a number of locations in Canada, abandoned coal tar waste sites are causing problems when the sites are redeveloped or where there is significant off-site movement of contaminants.

The Canadian Council of Resource and Environment Ministers (CCREM) Waste Management Committee was requested at the June 18 and 19, 1987 meeting of the Toxic Substances Advisory Committee to strike a working group whose responsibility would be to develop interim guidelines for PAH contaminants associated with abandoned coal tar sites.

The development of interim PAH guidelines was undertaken recognizing that several initiatives had already begun in this area. Specifically, Ontario is leading an initiative to develop multi-media standards for PAH under the general aegis of CCREM. Another ongoing initiative is the development of a risk-assessment-based methodology which will be used to establish action level criteria as called for under the CCREM Hazardous Waste Action Plan. However, these initiatives are multi-year undertakings which will not be completed in time to address the immediate environmental management problems posed by abandoned coal tar waste sites.

The interim PAH guidelines developed in this report are intended for use by the member governments of the CCREM until definitive criteria are developed.

1.1 APPLICATION OF THE INTERIM GUIDELINES

These guidelines have been developed for guidance in evaluating PAH contamination at coal tar waste sites to determine what, if any, remedial action may be required.

Decisions on the need to initiate such remedial work will be made by the appropriate agencies on a case-by-case basis.

Factors affecting such decisions would include current land-use and proposals to change land-use; any monitoring information showing the movement of contaminants off-site or into groundwater; or information on current environmental damage or suspected human exposure.

1.2 PAH WORKING GROUP

1.2.1 Terms of Reference

Terms of Reference were developed by the Ad Hoc Working Group and approved by the Waste Management Committee and the Toxic Substances Advisory Committee.

1. The Working Group shall review the problems associated with abandoned coal tar sites to determine for what media interim PAH guidelines are required, e.g. soil, water, sediment, or air, etc.

2. Based on a review of available information, the Working Group shall determine what guidelines are available for adoption from jurisdictions elsewhere.

3. The Working Group shall review initiatives underway by the member governments to determine whether new interim guidelines are required in the event that suitable criteria are not available for adoption.

4. The Working Group shall prepare a report to the Waste Management Committee of the Toxic Substances Advisory Committee under CCREM, recommending interim PAH guidelines including supporting rationale.

5. The Working Group shall recommend other contaminants associated with abandoned coal tar sites for which guidelines may be required.

1.2.2 Committee Membership

The Ad Hoc Working Group is comprised of a Chairman and one other member from Ontario plus representatives from Nova Scotia, Quebec, British Columbia and Environment Canada as nominated by the members of the Waste Management Committee. In addition to the main representatives, other technical experts from the member governments were consulted on an as-needed basis.

Committee Members

Beverley Hanna Thorpe, Chair, Ontario
Neil Brodie, Nova Scotia
Tom Foote, Environment Canada
Serge St. Laurent, Quebec (replaced M. Beaulieu)
Ulo Sibul, Ontario
John Ward, British Columbia

Technical Advisors

Brendan Birmingham, Ontario
Ken Mancuso, Health & Welfare Canada
Otto Meresz, Ontario
Mark Richardson, Health & Welfare Canada (formerly of Environment Canada)
Akos Szakolcai, Ontario

Secretary

Adam Socha, Ontario

1.3 APPROACH TO THE TASK

The prime approaches taken in this task were:

- to use information and guidelines developed elsewhere;

- to complete the task within 6 months;
- to keep the guidelines simple and pragmatic; and
- to reduce overlap with other on-going initiatives.

The Working Group held a series of teleconference meetings commencing at the end of September 1987. Members at the outset provided others with copies of pertinent technical materials and shared their experiences and approaches to the clean-up of coal tar sites within their respective jurisdictions. From this extensive information exchange, the scope and detailed approach to the development of interim PAH guidelines evolved. The information exchange had a particularly helpful spin-off benefit as much of this material was not readily available.

The Working Group identified the PAH of concern based on a review of information available in the literature and on the monitoring information from on-going remedial work in their jurisdictions. This list and rationale is found in Section 3.

Based on this list of PAH of concern and associated information on the environmental transport and exposure pathways of these PAH from abandoned coal tar waste sites; the important environmental media were identified where interim guidelines were needed.

While developing interim guidelines for contaminants other than PAH was outside the scope of this Working Group's responsibilities, it was requested to identify priority contaminants for further action to the Waste Management Committee. The Working Group prepared a detailed list of chemical classes of concern at coal tar sites which is included as Appendix 2.

The Working Group have recommended the selected list of contaminants found in Section 3 from the original list, based on their knowledge of which contaminants are present at reasonably high concentrations and/or which are most toxic. As new information becomes available, the list of compounds for future review may require revision.

Based on an extensive literature search, supplemented by information provided by Working Group members, a compilation of existing standards, guidelines or objectives, etc. for PAH developed or used elsewhere was prepared. This review formed the major basis for the identification of guidelines that could be recommended for adoption. This information and explanatory text is included as Appendix 1.

The guidelines available for adoption were then reviewed on the basis of appropriateness of the media and the match to our list of PAH, and on the applicability of the rationale and scientific basis upon which they had originally been developed. From this evaluation the interim guidelines recommended for adoption were developed. Section 7 sets out the recommended guidelines and supporting rationale.

To ensure that the Working Group's activities did not duplicate or contradict other CCREM initiatives, consultations were held with key personnel on: the Decommissioning Guideline Committee and its Working Group and their prime consultant, Monenco Ltd., the CCREM Task Force on Water Quality Guidelines, and also the Ontario Multi-Media PAH Guideline Development Committee.

The output of the decommissioning work will include a risk assessment-based methodology for identifying site-specific cleanup criteria for decommissioning contaminated industrial sites. The water quality group is preparing recommendations on the feasibility of developing sediment quality guidelines for Canada including recommended approaches for guideline development and selection of priority chemicals. The multi-media PAH guideline work is to develop guidelines for PAH for all exposure pathways to ensure that exposure from all media does not exceed allowable exposure based on human and environmental health considerations. This PAH guidelines report will be forwarded to the Multi-Media Guidelines Advisory Committee which reports jointly to CCREM and the Advisory Committee on Environmental and Occupational Health.

2.0 OVERVIEW OF COAL TAR WASTES IN CANADA; IMPORTANCE OF PAHS IN THESE WASTES

2.1 HISTORICAL OVERVIEW

2.1.1 Coal Gasification Industry in Canada

As early as the 1820's, much of the gas used for lighting, heating and some industrial processes was produced in coal gasification plants. These facilities were found in urban areas. Most of the larger cities, and some smaller towns across Canada had such plants until the introduction of natural gas and pipeline transmission drastically reduced their commercial viability. By the 1950's, most gasification plants were abandoned, demolished, or converted into other industries.

Coal gasification plants ranged in size from less than half an acre (.2 hectares) to over 100 acres (43 hectares) (Environmental Research and Technology, 1984). The main part of the site was the reactor building, where the coal was carbonized, carburetted, and otherwise converted to gas. Although it constituted the most important element of a gasification plant, the reactor building did not compose the greatest area of the site. Spread out in various locations were coal and coke stockpiles or sheds, new and spent oxide containers, gas containers (large metal cylindrical containers), tar wells, sludge ponds, and gas liquor reservoirs.

In larger plants, it was quite common to have distillation buildings and by-product recovery facilities on the site (Environmental Research and Technology, 1984). While these sites may be the source of additional problems, they would also indicate a lower likelihood of coal tar wastes remaining in storage containers on the site.

Many processes and types of plants were used to produce varied qualities of manufactured gas. The most common products are referred to as coke oven gas, retort gas, carburetted water gas, and producer gas.

2.1.2 Other Coal Tar Products

It is important to note that coal tar, which is the main substance of concern from coal gasification plants, is also produced and used in other types of industries.

Any operation which used gas produced from by-product coke or coal, for heating, smelting, or running equipment, also produced coal tar as a waste material. Examples of these are steel industries, city gas producers, mining operations and coking industries.

Other industries, such as coal tar distillers and by-product manufacturers, must also be considered when identifying sites which may be contaminated by coal tars. These companies, which produced ammonium sulphate, road tar, creosote, sealants, and/or wood preservatives, may have left coal tar in surface or underground storage containers upon site abandonment. Also, the storage facilities for the coal tar may have leaked on the site during or after plant operation.

2.1.3 Nature of Coal Gasification Plant Wastes

Waste and by-product materials from these processes were sometimes refined for other uses, but in all cases, at least some waste remained which required disposal. During the period of coal gas production, little concern was given to pollution and potential health effects resulting from these processes. For example, wastes were often stored on-site in underground containers, or surface sludge ponds. Discontinuation of a plant often meant demolition of buildings, which temporarily hid the stored wastes, or caused them to be spread out over a larger area. Little in the way of remedial measures were undertaken at these sites, so that at present, there are numerous possibilities for public health and environmental hazards to arise.

The characterization of coal gas plant wastes is complicated by the fact that a variety of production methods and many ranks or compositions of coal were used between the early 1800's and the mid-1900's (Mahlum et al, 1986). The main types of gas plants were coke oven, carburetted water gas, producer gas, and retort gas operation (Environmental Research and Technology, 1984). These produced a variety of wastes and by-products including, coal tar, sludge and pitch, ash and slag, sulphur, ammonia and other nitrogen compounds, wastewater, spent oxides, and numerous organic compounds.

Coke oven gas plants produced substantial by-product coal tar, however, most of this tar was recycled on-site or sold to coal by-product manufacturers and distillers such as Stelco in Hamilton, Ontario (Intera Technologies Ltd., 1986). The distilled tar was used to produce some industrial fuels, road and roofing tars, wood preservatives, and sealants (Braunstein et al, 1981). As such, most coke oven gas plant sites may be expected to have less coal tar wastes than coal gasification plant sites.

Carburetted water, producer, and retort gas plants tended to retain most of the coal tar on site, either in storage tanks, or sludge ponds. Water gas plants generally produced more tar in the coal gasification process, than did producer gas or retort gas plants (Environmental Research and Technology, 1984). Some water gas plants did recycle the coal tar wastes, thus lessening the likelihood of large coal tar reserves on abandoned plant sites. However, producer and retort plants, which produced approximately 10 gallons of dry coal tar per ton of dry carbonized coal, typically did not recycle the bulk of the waste tar material (Wilson and Wells, 1950).

The main coal gasification by-products of concern in this study are coal tars, sludges and pitches. Wastewater produced during plant operation might be of concern, however, dilution, degradation, and volatilization

processes which have been in operation since the waste water entered the environment (minimum 30 years) would have reduced the effluent to, at worst, a moderately toxic state.

Coal tar is comprised of numerous organic and inorganic compounds. The main chemical classes are polycyclic aromatic compounds (PAH), phenolics, light aromatics, nitrogen and sulphur compounds, and trace metals (Environmental Research and Technology, 1984). Relative to these classes, the main difference between the various coal gasification processes is that tars from carburetted water gas production contain no phenolics.

2.2 RELEVANT ISSUES, INITIATIVES AND ACTIONS IN CANADA

The assessment and management of contaminated sites containing coal tars have been addressed in a number of programs in Canada. Several issues have been considered.

Of primary importance is the development of coal tar site inventories, and work in several jurisdictions has addressed this specific need. Another important issue relates to standards and guidelines for these sites.

Significantly, a number of specific sites have been assessed and restoration and/or mitigation work has begun or has been completed.

2.2.1 National and Provincial Inventories

Under the direction of Environment Canada, the Resources Development Research Centre of Carleton University was recently contracted to review abandoned coal gas works in Canada. In the resulting report (Carleton University, 1987), 150 coal gasification plant operations were identified.

Eighty were recorded as being definite coal gas producers, thirty were listed as handling refined coal tar, and the remainder as possible coal gas producers. The sites were identified through archival research and direct contact with companies and government agencies. The report indicates

that abandoned coal gasification plant sites exist in all but one of the Provinces (Prince Edward Island), and that Montreal, Toronto and Vancouver have the greatest number of sites per city. It should be noted that the research involved in this report apparently did not involve visits to the abandoned sites, so their location and existence was not directly confirmed.

2.2.2 Ontario

The Ontario Ministry of the Environment has also developed an inventory of manufactured gas plant sites. In a phase one program, Intera Technologies Ltd. was contracted to identify and provide a preliminary assessment of "town gas" facilities that manufactured coal gas for municipal illumination and heating. The resulting report listed 41 manufactured gas plants in 36 different communities in Ontario (Intera Technologies Ltd., 1987). Industrial facilities and off-site disposal areas for gas plant wastes were not included.

In contrast with the National Overview carried out by Carleton University, site reconnaissance visits were made to identify any visual or olfactory evidence of buried waste. Sewers, basements, and sumps were also inspected. A qualitative assessment of the potential environmental impact of the sites was performed, and options for further actions were developed.

Since the initial municipal coal gasification plant site inventory was completed, Ontario is undertaking investigations at all coal gasification plant sites to determine the presence and extent of waste on each site. These studies will also focus on off-site contamination of soil, streams, lakes, ground water, and in some cases, indoor air quality. Appropriate remedial actions will be taken where unacceptable levels of contamination are found. To date, cleanups in Rideau River (Ottawa), Chippawa Creek (Niagara Falls) and at Waterloo have been completed. Several other sites are presently under consideration.

To complete the inventory of the major sources of coal tar waste in the Province, industrial sites which produced or used coal tar, are being identified. Processes and uses include coal tar distillation, water proofing, coke ovens, roofing materials and pinstch gas plants.

2.2.3 Quebec

In 1984, the Ministere de l'Environnement du Quebec announced an inventory of sites used for the disposal of hazardous wastes in Quebec. This was updated in 1987, and was developed by the GERLED (Groupe d'etude et de restauration des lieux d'elimination de dechets dangereux) Task Force, which used a number of means to identify, investigate and cleanup the sites. Included were review of government files, input from citizen's environmental groups, surveys of municipalities, scrutiny of aerial photographs, and a review of the industrial profile of each Ministry region. In 1985, various criteria were used for the cleanup of the Ville LaSalle landfill. The earliest version of the "ABC" criteria (see Appendix 1) were used in the investigation and cleanup of soil and groundwater. The WHO criteria were used for drinking water.

PAH in air were not analysed, but, rather benzene, toluene and xylene were monitored.

The production of coal tar gas in Quebec ceased in the early 1970's. An inventory of coal tar waste sites was carried out during the summer of 1987 by Environment Quebec. A total of 26 sites in the province had been used for the production of coal gas and/or the refining of coal tar.

Two of the sites in the GERLED inventory of contaminated sites were Quebec Power and LaSalle Coke. The investigation of these sites is complete. The cleanup of Quebec Power's land in Quebec city will start in October 1988. During the summer of 1988, 480,000 m^3 of contaminated soil from the LaSalle Coke site will be buried in a maximum security

landfill installed on site. Additionally, two sites in Montreal have shown evidence of contamination of soil and groundwater based on preliminary analyses. The remaining two sites are located on federal properties (McGill Street in Montreal and the Canadian Coast Guard installation in Sorel); the investigaton of these sites will be the responsibility of the federal government. Consideration is being given to the establishment of a special fund to investigate the 20 other coal tar sites in Quebec by the end of 1989 and to determine the need for intervention on these sites.

2.2.4 Other Provinces

Activity concerning coal tars in other Provinces has usually centred on the restoration of specific problem sites.

For example, in Nova Scotia, the contamination of Muggah Creek and the Sydney Tar Ponds adjacent to Sydney Steel Corporation has posed a considerable challenge. The cleanup operation is expected to take 10 years to complete and cost $50 million for the 37 hectare site.

In British Columbia the contamination of the former EXPO 86 site has been extensively studied. While a risk assessment has been carried out, no definite plans for mitigation or restoration have been developed.

Alberta has inspected their one "probable" site and found no evidence of hydrocarbon contamination. The sites listed as "possible" have been researched with no evidence of coal gasification plants being found.

2.2.5 CCREM Initiatives

Abandoned coal tar sites were discussed at the June 18 and 19, 1987 meeting of the CCREM Toxic Substances Advisory Committee (TSAC). Some jurisdictions had immediate need for guidelines for coal tar sites. It was determined that a

small working group, reporting to the CCREM Waste Advisory Subcommittee, be created to recommend criteria for abandoned coal tar sites in Canada. It was further decided that since Ontario had already made considerable progress in developing criteria for PAH, and that these coal tar components were among the most hazardous, that the Working Group should first focus on these materials.

In a broader context, the activity of the Coal Tar Working Group fits into the general contaminated sites activities under the CCREM Hazardous Waste Action Plan, which is the main focus of the Waste Subcommittee. Action item 4, of the Hazardous Waste Action Plan which involves decommissioning requirements, has a number of activities that will address coal tar sites. For example, action item 4.1 involves the development of action level criteria and criteria for cleanup of contaminated sites.

Nevertheless, TSAC felt a fast track approach would be necessary for these particular wastes.

3.0 LIST OF CONTAMINANTS OF CONCERN

The major types of waste that may be found at old coal gasification plant sites are tars and sludges, spent oxides and ash, slag and clinkers.

Environmentally, tars and sludges present the greatest concern because of their physical mobility and the ability to contaminate soil, water and air with hazardous organics at and near former gas plant sites. The major contaminants of tars are mainly PAH.

The contaminants contained in solid wastes such as spent oxides and various ash and slag are generally considered to be of less concern because of their low bioavailability.

For the purpose of developing interim guidelines, the PAH can be divided into two general groups. The substances in Group 1 (Table 3.1) consist of PAH designated as carcinogenic by the International Agency for Research on Cancer (IARC); the substances in Group 2, although not documented as being carcinogenic but are good indicators of coal tar contamination. Section 4 of this report discusses the environmental and health hazards of PAH in more detail.

Two additional groups of contaminants (Group 3 and Group 4) are identified as substances of concern at coal tar waste sites. These contaminants should be considered candidates for future review in developing interim guidelines. These other contaminants occur commonly in high concentrations in soil or water contaminated by coal tars. Therefore the presence of these contaminants may restrict the development of land for some uses.

3.1 PAH

The polycyclic aromatic hydrocarbons in coal tars are considered generally of greatest concern because some are carcinogenic and consequently this group of chemicals are of prime concern in the cleanup of old gas plant sites.

3.1.1 Group 1: Carcinogenic PAH

The six substances in this group (Table 3.1) are considered by the IARC to be carcinogenic, with dibenz[a,h]anthracene and benzo[a]pyrene (B[a]P) being the most potent carcinogenic PAH. These six substances are common constituents in coal tars and can be found in contaminated soil, water and air. Drinking water and ambient air criteria have only been developed for B[a]P (Appendix 1). B[a]P is often used to gauge the hazardous nature of contaminated soil.

These substances are also often used in risk assessment models. Routine analytical methods are available for these compounds.

3.1.2 Group 2: Other PAH

Naphthalene, phenanthrene and pyrene make up this group. They are usually good indicators of contamination by coal tars because they are often found in high concentrations in contaminated soil, air and water.

These compounds are 2, 3 and 4-ring structures respectively, in contrast to the Group 1 PAH which are mostly 5-ring structures. Due to their smaller size, these PAH compounds are more mobile and will move more quickly into groundwater and volatilize into air from soil.

Compared to other PAH, naphthalene is the most soluble and volatile and gives coal tar contaminated soils a distinctive odour. Because of its volatility, naphthalene may be present in air where coal tar contamination is suspected.

All three compounds, because of the usually high concentrations found in soil contaminated with coal tars, would be good indicators of the effectiveness of the specific cleanup efforts.

3.2 GROUP 3: OTHER ORGANIC COMPOUNDS

Of the other organic compounds commonly found in coal tars, benzene, toluene, and xylene (BTX) should be considered for future reviews for the establishment of guidelines because they are principal constitutents of coal tars. They have been found to be present in soil, air, and water that has been subjected to contamination by coal tar.

BTX are present throughout the environment as a result of various man-made sources (Environmental Research and Technology, 1984). They have a very low tendency for adsorption, and may dissolve in groundwater. The main form of attenuation of BTX, besides their volatility, is biodegradation, which is very effective in aerobic zones in the soil.

Benzene is carcinogenic and neurotoxic. BTX are moderately persistent and volatile. When present in ambient air at coal tar waste sites, they likely indicate fresh contamination by coal tar. As with the substances in Group 2, BTX may be used to conduct post-cleanup monitoring where fresh coal tars have been encountered. Because of their volatility, they may be present in air.

3.3 GROUP 4: INORGANIC COMPOUNDS

Coal tar contains numerous inorganic compounds, including iron, ammonia, cyanide, and cyanide complexes, nitrate, sulphate, elemental sulphur, arsenic, chromium, lead, and zinc (Braunstein et al, 1981). Certain nitrogen and sulphur species are biodegradable to some degree and most metals adsorb to soils, thus limiting the potential adverse effects of these compounds.

These inorganic substances are associated with by-products or processes of coal gasification and/or impurities in the coal. Spent oxides are a major source of these substances and very large concentrations of these substances can be found in contaminated soil and water.

The presence of these inorganics in soil at Canadian coal tar plant sites has not been determined routinely, but most of the substances have been found in large concentrations in soils at plant sites in the United Kingdom (Wilson and Stevens, 1981).

Future consideration should be given to conducting analysis of, and setting guidelines for, the substances in this group. All have established drinking water objectives and may play a significant role where surface or groundwater contamination is likely by coal tars. Sulphide/sulphates may also affect concrete structures.

If present in large concentrations, the substances may also be indicators of an old gasification plant and tracers of both possible coal tar contamination and of other types of contamination associated with coal gasification plants. In this context, they may be used to define the areal extent of contamination at a site, as well as for post cleanup monitoring.

TABLE 3.1: LIST OF SUBSTANCES FOR CONSIDERATION FOR THE DEVELOPMENT OF CLEANUP CRITERIA

GROUP 1: Carcinogenic PAH

benzo[a]anthracene
benzo[b]fluoranthene
benzo[k]fluoranthene
benzo[a]pyrene
dibenz[a,h]anthracene
indeno[1,2,3-cd]pyrene

GROUP 2: Other PAH

naphthalene
phenanthrene
pyrene

GROUP 3: Other Organic Compounds (for future review)

benzene
toluene
xylene

GROUP 4: Inorganic Compounds (for future review)

iron
arsenic
sulphide/sulphate
iron-cyanide complexes
free cyanide

4.0 HAZARDS ASSOCIATED WITH PAH

The presence of PAH from coal gas plant sites in air, water or soil at concentrations above natural background levels may affect human health, plant or animal life. Assessment of whether these effects are adverse and significant depends on the toxicity of the specific PAH or mixture of PAH, the exposure level, the exposure pathway(s) and the exposed organism or population.

While human health impacts are generally those of most concern at waste sites, aquatic and terrestrial impacts must also be considered. Gas plant waste constituents can affect aquatic ecosystems via surface run-off or groundwater discharges to nearby surface waters. Terrestrial life can be affected by direct contact with wastes, contaminated soils or volatilized constituents, or by contaminated surface water run-off and shallow groundwater contamination.

4.1 PHYSICAL-CHEMICAL PROPERTIES OF PAH

The hazards associated with PAH and the movement and behaviour of PAH in the environment are dependent on their physical properties. The physical properties of PAH are dependent on the structure of individual compounds (Table 4.1). The aqueous solubility of each compound decreases as the molecular weight of the compound increases. Therefore those compounds with a simple structure have higher solubilities than those with a complicated structure. A similar relationship exists for volatility of PAH whereby the boiling point temperature increases with increase in molecular weight. PAH are strongly adsorbed to soils and tend to be relatively immobile in the environment. PAH are also susceptible to biodegradation by microorganisms.

TABLE 4.1 : PHYSICAL PROPERTIES OF COMMON COAL TAR PAH

	CAS NUMBER	MOLECULAR WEIGHT	BOILING POINT (°C)	VAPOUR PRESSURE (mm Hg)	AQUEOUS SOLUBILITY mg/L @ 25°C
Naphthalene (2-ring)	91-20-3	128.2	218	0.082	34
Phenanthrene (3-ring)	85-01-8	178.2	338	6.8×10^{-4}	1.6
Pyrene (4-ring)	129-00-0	202.3	393	6.75×10^{-6}	0.16
Benzo[a]anthracene (4-ring)	56-55-3	228.3	435 *	5×10^{-9}	0.015
Benzo[b]fluoranthene (5-ring)	205-99-2	252.3	481	(10^{-11} to 10^{-6})	0.0015
Benzo[k]flouranthene (5-ring)	207-08-9	252.3	481	9.5×10^{-11}	0.0008
Benzo[a]pyrene (5-ring)	50-32-8	252.3	496	5×10^{-8}	0.004
Indeno[1,2,3-cd]pyrene (6-ring)	193-39-5	276.3	534 **	1×10^{-10}	0.078
Dibenz[a,h]anthracene (5-ring)	53-70-3	278.4	524 **	(10^{-10} to 10^{-14})	0.0005

* sublimates
** estimated

4.2 MOVEMENT IN THE ENVIRONMENT

Freshly exposed coal tar and coal tar contaminated material is easily identifiable when encountered because of its distinctive odour which resembles creosote. Weathering diminishes the odour substantially.

The tar is typically a brown to black resinous material with a consistency varying from a light oil to a heavy tar. It is immiscible with, and denser than, water and therefore exhibits density-dependent flow characteristics. When in contact with surface water, coal tar may release a phase which causes an oily sheen on the water surface. Immiscible coal tar in surface water will sink to the bottom of the water body and will collect in depressions.

However, the fact that PAH occur in tars also affects their environmental mobility. The BTX content of the tar influences its odour properties and may contribute to the release of PAH in a mobile lighter-than-water phase to water in contact with freshly exposed coal tar. The major component of tars is an immiscible, denser-than-water fraction which is relatively immobile in soil but may migrate over limited distances in porous surficial sediments following discharge or erosion into the aquatic environment.

4.3 ROUTES OF EXPOSURE AND ABSORPTION

Humans are exposed to PAH of all types during their entire lifespan. PAH are ubiquitous in the environment. PAH are found in polluted air, tobacco smoke, cooking products, soots, tars and oils. PAH are formed by a variety of combustion and incineration processes routinely exposing most people to low levels of PAH. In general, PAH are a large group of chemicals of which only a small fraction have been tested for human health effects. The major routes of PAH exposure are through inhalation of polluted air, through ingestion of PAH-containing food, through dermal contact with materials rich in PAH and in some cases, through ingestion of PAH-contaminated dusts and soil.

From the perspective of coal tar plant sites exposure pathways will be determined by the proposed use of these sites. Exposure scenarios during cleanup of the sites will differ from the typical exposures that will occur after cleanup depending upon whether industial, residential housing or agricultural or parkland uses are involved.

In the residential housing use pattern, inhalation exposure will be related to volatile or dust-borne PAH derived from the cleaned up site, ingestion of PAH on home-grown fruit or vegetables, dermal contact with and ingestion of soil associated with backyard recreational activities and indoor dust derived from on-site soils.

As with all chemicals, it may be assumed that biological effects will result from exposure to PAH if the exposure is of sufficient intensity and duration. Regardless of the route of absorption by humans, exposure to PAH always occurs in the form of a complex mixture rather than a single compound. This fact complicates the evaluation of PAH toxicity to humans, since different PAH sources produce different exposure profiles.

4.4 HUMAN TOXICITY

The health effect of major concern for PAH is the potential for development of cancer.

Given our concern with coal tar wastes this review will be restricted to nine PAH common to the complex mixtures associated with coal tar sites. Six of these PAH are designated as carcinogenic based on their classification by IARC. The other three PAH are not carcinogenic but are indicators of coal tar contamination due to their presence at high levels at these sites.

Toxicologic data are not available for all the PAH known to exist, however, the nine coal tar waste PAH are well studied. Most of our understanding concerning the toxic effects of PAH is derived from studies with experimental animals exposed to a single PAH. In general, it may be concluded that the acute poisoning potential of PAH by any route of absorption is relatively low, such as benzo[a]-pyrene, phenanthrene, and pyrene. The acute LD_{50} (single dose required to kill half the animals) falls in the range of 250-1000 mg/kg of body weight (Table 4.2). In addition, there exists limited evidence that some PAH (e.g. benzo[a]pyrene) produce adverse effects on fertility and on the developing fetus in experimental rodents.

TABLE 4.2 : TOXICITY OF COMMON COAL TAR PAH

	ACUTE TOXICITY LD_{50}*	MUTAGENICITY** (ANIMAL OR BACTERIAL)	CARCINOGENICITY*** (DEGREE OF EVIDENCE FOR ANIMALS)	REPRODUCTIVE EFFECTS
Naphthalene	1 g/kg bw (rat, i.p)	0	No evidence.	
Phenanthrene	700 mg/kg bw (mouse, i.p)	0/+	Inadequate.	
Pyrene		0/+	No evidence.	
Benzo[a]anthracene		++	Sufficient.	
Benzo[b]fluoranthene		+	Sufficient.	
Benzo[k]fluoranthene		+	Sufficient.	
Benzo[a]pyrene	250 mg/kg bw (mouse, i.p)	++	Sufficient.	60 mg/kg(rat, i.p.)
Indeno[1,2,3-cd]pyrene		+	Sufficient.	
Dibenz[a,h]anthracene		0/+	Sufficient.	

* LD_{50} - the lethal dose which kills 50% of the test animals; i.p. - interperitoneal injection

** - 0 - no mutagenicity
- + - moderately mutagenic
++ - strongly mutagenic

*** - Based on the evaluation by the International Agency for Research on Cancer (IARC) of published literature on cancer studies.

Reported cases of acute PAH poisoning in humans are virtually nonexistent. The only major exceptions are for naphthalene, which is known to produce hemolysis (destruction of red blood cells) and cataracts in humans (ACGIH, 1983), and numerous case reports of phototoxic reactions of the skin associated with exposure to petroleum pitch (Bingham et al, 1980).

The carcinogenic properties of PAH and their simple derivatives have been studied in animals for more than 50 years, and their role in the causation of cancer in humans was first implied more than 200 years ago. The accumulated information concerning the carcinogenic potency and mechanism of action of PAH indicates that carcinogenic PAH have several features:

1. PAHs such as benzo[a]pyrene are powerful carcinogens and mutagens;
2. tumors can be produced in laboratory animals by a single exposure;
3. they act both at the site of contact (e.g. skin, lungs) to produce tumors, as well as at organs distant to the site of absorption; and
4. their carcinogenic effects have been demonstrated in nearly every species tested, regardless of the route of administration.

The more abundant PAH in the environment (e.g. pyrene, phenanthrene, naphthalene) are not carcinogenic however, PAH are generally found in the environment as complex mixtures containing both carcinogens and noncarcinogens. Thus the problem arises of how to evaluate the carcinogenic risk to humans from different types of PAH-containing substances.

One approach to assessing human cancer risks from PAH involves animal bioassay studies conducted with actual PAH-containing substances, or extracts derived therefrom. The available bioassay data on complex PAH mixtures is largely restricted to the results of mouse skin-painting and subcutaneous injection studies with extracts of tobacco tars, coal tars and pitches, coke oven emissions, gasoline and diesel engine exhaust, etc. Dermal application of coal tar to mice results in increases in dermal tumours and animals exposed to coal tar aerosols develop lung tumors (U.S. EPA, 1984).

Human cancer risk can also be assessed from humans exposed to high levels of coal tar. Epidemiological studies of roofers and coke oven workers who are exposed to coal tars or coal tar volatiles indicate an increased risk of skin, lung and kidney cancers. Human case reports dating back to 1885 support an association between high occupational exposure to coal tars and increases in skin cancer.

The quantitative effect on tumour response resulting from interactions among individual PAH in the mixture, both carcinogenic and noncarcinogenic, is not understood.

A number of cancer risk assessment approaches have been developed to tackle this problem of complex PAH mixtures. One approach has been to develop some relationship between the benzo[a]pyrene content of a mixture and its carcinogenic potential. Typically B[a]P is a minor constituent of most mixtures and this approach ignores the contribution of the other carcinogenic PAH. Recently relative potency ranking schemes have been proposed which take the slope of the cancer bioassay data for the individual PAH and ranks it in comparison to that of B[a]P. Since there are a wide variety of animal species, exposure routes, target organs and experimental protocols used in cancer bioassays it may not be prudent at this time to accept this risk assessment methodology.

4.5 EFFECTS ON AQUATIC ECOSYSTEMS

A great deal of research has been conducted with PAH and their effects on aquatic life in recent years. Acute, chronic and bioconcentration studies have been conducted on many of the PAH compounds. However, the majority of these studies have been conducted with marine (salt-water) species as a result of oil spill impact studies (Neff, 1979).

In general the acute toxicity of PAH to a particular species increases as the compound molecular weight increases. However, low acute toxicity of high molecular weight compounds (for example, chrysene, and benzo[a]pyrene) is probably due to low bioccumulation potential since alkyl substitution increases acute toxicity. On the basis of acute toxicity testing, aquatic crustaceans (shrimp, etc.) are most sensitive, polychaetes (aquatic [mostly marine] worms) are intermediate in sensitive and fish least sensitive of the groups tested. Chronic exposures to low concentrations of PAH in water, food or sediments may produce sublethal responses in aquatic organisms. These effects can include, but are not limited to , reduced survival, behavioural changes, impaired reproduction, or, in some cases, possible cancer induction in sensitive species. In most cases, PAH concentrations required to elicit significant chronic responses are higher than those

concentrations normally encountered in all but the most heavily polluted aquatic environments.

As in mammalian studies, it has been shown that fish have some of the same enzymatic systems which attack PAH molecules causing breakdown into metabolites. The majority of PAH metabolites breakdown to form phenols or are converted by enzymes to form glutathione, glucuronide or sulphate conjugates which are rapidly eliminated from the organism. Certain PAH metabolites are known to be mutagenic, and carcinogenic to the host organism. Only a few aquatic organisms have been studied extensively in regards to carcinogenicity. In those studies, induction of cancer by exposure of aquatic organisms to environmentally realistic levels of PAH has not been reported. In general, field investigations have not definitively identified specific PAH compounds as the causative agent in aquatic populations with increased incidences of cancer.

In contrast, PAH are known to accumulate in tissues of aquatic invertebrates as a result of exposure from water, food or sediments. This is not surprising because of the highly hydrophobic and lipophilic nature of the PAH compounds and the fact that invertebrates are generally unable to metabolize PAH. Both abiotic and biotic factors affect the rate of PAH uptake, retention and depuration. In general, accumulation from water is more important than uptake from food or sediment due to the adsorptive chemistry of PAH compounds. Most aquatic organisms demonstrate the ability to metabolically degrade and excrete PAH.

4.6 EFFECTS ON TERRESTRIAL ECOSYSTEMS

In the terrestrial environment, effects on animal life and plant life, including both direct toxicity and food-chain impacts, are of concern. The toxic effects of PAH on mammals can be inferred from the extensive toxicity testing work performed on laboratory animals. As with humans, the basic conclusion is that PAH are only slightly to moderately toxic by acute exposures, but chronic exposures to certain PAH compounds can result in cancer. Bioconcentration in

animal food chains is unlikely since PAH are readily metabolized. Raw meat does not normally contain PAH, and reported PAH concentrations in meats usually originate from the cooking process itself.

The fate and impacts of PAH in terrestrial plant systems have recently been reviewed by Santodonato et al (1981) and Edwards (1983). These reviews conclude that the principal mechanism of PAH uptake by plants is by adsorption rather than absorption. In other words, PAH will accumulate on the surface of roots and leaves but very little PAH will be translocated from the roots to the shoots. Thus for crops grown in contaminated soil, PAH concentrations are likely to be higher in the roots than in above-ground plant parts. It has also been reported with some plants that PAH translocation to other plant parts can occur, with the translocated PAH possibly accumulating in plants grown in highly contaminated soils, where the rate of uptake exceeds the rate of metabolism and degradation (Edwards, 1983).

Data on the phytotoxicity of PAH are limited. Wilson and Stevens (1981) report that small scale tests with a commercially available coal tar showed inhibition on the growth of radishes and grass. Weathering of the contaminated soil diminished the phytotoxic response which suggests that volatile, non-PAH components of the coal tar are involved in the phytotoxic responses observed. Phytotoxicity tests have also been conducted with samples of various coal-derived synthetic fuel oils and tars, which are roughly similar to coal tar in composition (i.e., highly concentrated in PAH). Such tests have also shown growth inhibition effects (Cline et al, 1981).

5.0 REVIEW OF BACKGROUND LEVELS OF PAH WITH EMPHASIS ON CANADIAN DATA

5.1 AMBIENT AIR

Measurement of urban ambient air in Canada for PAH would appear to date from the early 1970's (Adamek, 1976). Most of the early ambient air data come from Ontario (MOE, 1979; Katz et al, 1978; Pierce & Katz, 1975, Potvin et al, 1981). More recently, other information on current levels of PAH in urban or industrialized areas across Canada have become available (Dann, 1988; Hoff and Chan, 1987).

Due to changes in the sampling and analytical approaches used and the types of PAH analysed for, there are problems with comparing earlier measurements of PAH in air with the most recent data sets. The recent literature indicates that sampling methods have to take into account volatility and reactivity of PAH.

For instance, earlier methods for monitoring ambient airborne PAH utilized flat porous filters only which collected the non-volatile PAH which are particulate bound but failed to sample the volatile, low molecular weight PAH in the vapour phase. Modern sampling techniques employ filters for particle-bound PAH in conjunction with absorbents downstream of the filter to trap vapour phase PAH. Other factors affecting sampling include the effect of ambient temperature on the partitioning of PAH between the vapour and particulate bound phases and reactions ocurring between PAH trapped in the sampling device and other gaseous reactants e.g. ozone, nitrogen and sulphur oxides passing through the sampler (Davis et al, 1987).

Of the coal tar-related PAH being considered in this report, napthalene, with the lowest molecular weight, is the most volatile, followed by phenanthrene and pyrene in order of decreasing volatility. These PAH are mainly found in the vapour phase. Benzo[b]fluoranthene and benzo[a]anthracene are intermediate in that they are found both in the vapour phase and bound to particulate. The remainder of the PAH under consideration are larger, non-volatile and particle bound.

Ranges of mean ambient air levels of the nine PAH under review are reported in Table 5.1. The data are selected from recent data sets for Montreal, Toronto and the Niagara region (Dann, 1988; Hoff and Chan, 1987). Other recent ambient air data from Kitimat, B.C.; Whitehorse, Y.T. and Sydney, N.S. are not included since they were collected near an aluminum smelter, in an area with large numbers of woodstoves and near coke ovens, respectively.

TABLE 5.1 : MEAN URBAN AMBIENT AIR LEVELS OF SELECTED PAH

Compound	Range of Reported Mean Values (ng/m^3)
Naphthalene *	3.4 - 70.2
Phenanthrene	5.0 - 15.6
Pyrene	0.37 - 5.1
Benzo[a]anthracene	0.4 - 2.8
Benzo[b]and[k]fluoranthene	1.3 - 3.3
Benzo[a]pyrene	0.2 - 0.6
Benzo[1,2,3-cd]pyrene	0.5 - 1.0
Dibenz[a,h]anthracene	0.04 - 0.12

* - Naphthalene not reported by Dann, 1988. Upper range value from background monitoring at a proposed incinerator site at Malton (Report to Petrosun/SNC Hearing, 1987)

5.2 BACKGROUND LEVELS IN SOIL AND SEDIMENTS

Information on Canadian background levels of PAH in soil from uncontaminated sites is scarce (Zenon, 1987; Hites et al, 1980; Wang and Meresz 1982). Most literature values are based on older U.S. and European Studies (Grimmer, 1983; NRCC, 1983). Some data on levels of PAH in sediments of the Great Lakes are available (Hites and Gschwend, 1982; NRTC, 1984; Eadie et al., 1982).

Urban background soil values from two Ontario studies are presented in Table 5.2 (Zenon, 1987; MOE, 1987).

TABLE 5.2 : URBAN SOIL BACKGROUND LEVELS OF SELECTED PAH

Compound	Mean	Range - (mg/kg)
Naphthalene	ND(0.03)	-
Phenanthrene	0.093	ND(0.1) - 0.95
Pyrene	0.11	ND(0.1) - 0.86
Benzo[a]anthracene	0.04	ND(0.1) - 0.17
Benzo[b]and[k]fluoranthene	0.054	ND(0.1) - 0.28
Benzo[a]pyrene	0.028	ND(0.005)- 0.18
Indeno[1,2,3-cd]pyrene	ND(0.03)	-
Dibenz[a,h]anthracene	ND(0.03)	-

Because of their low water solubility and hydrophobic nature, PAH tend to be associated primarily with inorganic and organic material in suspended and bed sediments (NRCC, 1983). Concentrations of benzo[a]pyrene in freshwater sediments range from 0.001 to 17 ug/g (dry weight) (Neff, 1979). Several PAH were identified in surficial and age-dated sediments from the Kitimat Arm downstream from an aluminum smelter in British Columbia (Cretney et al, 1982). PAH concentrations in sediments in Sydney Harbour, Nova Scotia, were found to be 10^3 - 10^4 times higher (up to 2829 ug/g (dry weight) for 12 PAH)than those found in the water column (Environment Canada, 1982).

5.3 PAH IN WATER

PAH are not uniformly distributed throughout the aquatic environment; instead, higher concentrations are generally encountered near point sources. The majority of PAH entering the aquatic environment are localized in rivers, lakes, estuaries and coastal marine waters. Concentrations of PAH in aquatic ecosystems are generally highest in sediments, intermediate in aquatic biota and lowest in the water column (Neff, 1979; NRCC, 1983).

River water may contain significant concentrations of PAH (0.02-3.79 ug/L total PAH) (Neff, 1979); in general, there is a direct relationship between the concentration of PAH in river water and the degree and nature of industrialization and/or urbanization in the specific watershed (NRCC, 1983). Groundwater usually contains PAH at concentrations approximately one order of magnitude lower than those of surface waters (0.0009-4 ug/L total PAH) (Neff, 1979), except in areas of specific localized contamination (NRCC, 1983).

6.0 ANALYSIS OF PAH IN ENVIRONMENTAL SAMPLES

Owing to their ubiquitous occurrence and environmental significance, the analysis of polycyclic aromatic hydrocarbons (PAH) has experienced phenomenal development during the pat fifteen years. A number of analytical techniques are now available for the analysis of PAH in various matrices (air, water, soil, fish, etc.). The important steps in each method are sampling, sample storage, pre-concentration (extraction), clean-up, separation and detection. Gas chromatography (GC), capillary column gas chromatography (CCGC) and high performance liquid chromatography (HPLC) are mostly used for the separation of components. The detection and quantification of PAH is achieved by using a flame ionization detector or mass spectrometer in combination with GC or CCGC and ultraviolet (UV) or fluorescence detector with HPLC.

Detection limits as low as 25 pg (10^{-12} g) injected can be achieved for certain PAH using HPLC and fluorescence detection. Regardless of the analytical method used, sample complexity can seriously affect method detection limits for PAH. It should be noted that although coal tar is one of the major sources of PAH it is also one of the most complex samples for PAH analysis.

Different analytical methods are used for different matrices. For example, the analysis of drinking water for PAH requires the lowest detection limits.

Table 6.0 summarizes the routine detection limits for the PAH considered in the present report. Lower detection limits can sometimes be achieved using state-of-the-art procedures and equipment.

TABLE 6.0 : METHOD DETECTION LIMITS

	MEDIUM		
	Water ug/L	Soil/Sediment mg/kg	Air ng/m^3
Benzo[a]anthracene	0.02	0.4	1.2-2.3
Benzo[b]fluoranthene	0.01*	0.3	1.2-2.3
Benzo[k]fluoranthene	0.001	0.2	1.2-2.3
Benzo[a]pyrene	0.005	0.2	1.9
Dibenz[a,h]anthracene	0.01	0.2	1.2-2.3
Indeno[1,2,3-cd]pyrene	0.02	0.3	1.2-2.3
Naphthalene	0.01	0.3	2.3
Phenanthrene	0.01	0.1	1.2
Pyrene	0.02	0.1	1.2
Total number of PAH and related compounds in scan	(17)	(16)	(41)

* - together with perylene

6.1 ANALYTICAL METHODS

The reliable detection of trace quantities of individual PAH in different environmental media requires a high degree of care in both the sample collection and handling and in the analytical procedures and equipment. At present, comparability of results from different workers cannot be ensured. There is a strong need for development of standardized analytical protocols for various media, including minimum quality assurance/quality control procedures. In conjunction with this, a series of inter-laboratory comparison round robins for both government and private labs and ongoing performance management and data review programmes would greatly enhance the reliability and comparability of PAH analyses.

PAH in Drinking Water

PAH in drinking water is analysed by reverse phase HPLC after solvent extraction from the sample followed by drying and dissolution in acetonitrile. Detection limits for individual PAH are in the low ng/L range (parts per trillion).

PAH in Sediments and Soil

Wet sediments are spiked with labelled PAH (to provide measure of the efficiency of the analytical procedure), extracted with solvent and concentrated. The extract is separated from interference chemicals and the resultant PAH containing fraction is analysed using GC-MS. Detection limits for the various PAH analysed in the wet sediments are below the part per million range.

PAH in Ambient Air

PAH in air are collected by Hi-Vol samplers using a filter usually backed up by resin cartridges or PUF (polyurethane foam) plugs. Due to the semi-volatile nature of many PAH in air, filters alone do not capture all the PAH in such samples. Considerable care and expertise are required in the design of sampling equipment and the sample collection and handling procedure to ensure an accurate result. Only recently has it been possible to collect reliable data on PAH in air.

The PAH on filters and back-up cartridges are extracted with methylene chloride and concentrated by evaporation followed by removal of interfering contaminants, i.e. cleanup. After further concentration, including solvent exchange, the sample is analysed by dual capillary column, dual flame ionization detection gas chromatogrpahy.

7.0 RECOMMENDED GUIDELINES FOR PAH

From the Working Group's review, it became clear that the principal media subject to contamination by coal tar wastes are soils in the immediate vicinity of the coal tar sites, and the groundwater. Sediment and surface water may also be of concern when the abandoned plant sites are located adjacent to waterbodies.

Air contamination is of lesser general concern due to the relatively low volatility of the carcinogenic PAH and is dependent on the age of the wastes. Potential air contamination can be of concern in some site-specific situations, e.g. in the excavation of coal tar wastes from enclosed storage tanks or where wastes are or might be enclosed in buildings constructed on top of the site. Such concerns may be more important from an occupational health perspective than for general environmental health. As such the agencies responsible for occupational health must take the lead.

The guidelines available for adoption were reviewed on the basis of appropriateness of the media and the match to our list of PAH, and on the applicability of the rationale and scientific basis upon which they had originally been developed. From this evaluation, the interim guidelines recommended for adoption were developed.

7.1 SOIL AND GROUNDWATER GUIDELINES

Upon review of the existing guidelines for PAH in soils and groundwater, the Working Group determined that the Quebec approach was the most suitable for adoption. These guidelines were found to be comprehensive, with a well-documented rationale for their application covering the primary PAH of concern applicable to coal tar waste.

7.1.1 Discussion of Recommended Guidelines

The Quebec Ministry of the Environment's "ABC" approach is a modification of a similar index developed by the Ministry of Housing, Physical Planning and Environment of the Netherlands.

The values for A, B and C have not been determined with a risk assessment model. In formulating the approach, it was clear that a lot of research remained to be carried out into the toxicological effects of substances to which the environment and human beings are exposed. The values are based upon the knowledge of different properties such as toxicity, from the Registry of Toxic Effects of Chemical Substances (RTECS), bio-accumulation potential, solubility in water, vapour pressure, viscosity, etc.

The "ABC" approach is considered by the Working Group to provide an effective tool for the management of coal tar waste sites. A, B and C values are presented for nine (9) PAH generally found at such sites (Table 7.1)

TABLE 7.1 : "ABC" VALUES* FOR PAH IN SOIL AND GROUNDWATER AT COAL TAR WASTE SITES

PAH	Conc. in Soil (mg/kg dry weight)			Conc. in Ground*** water (ug/L)		
	A	B	C	A	B	C
Group 1 Carcinogenic PAH**						
- benzo[a]anthracene	0.1	1	10	0.01	0.1	1
- benzo[b]fluoranthene	0.1	1	10	0.01	0.1	1
- benzo[k]fluoranthene	0.1	1	10	0.01	0.1	1
- benzo[a]pyrene	0.1	1	10	0.01	0.1	1
- dibenz[a,h]anthracene	0.1	1	10	0.01	0.1	1
- indeno[1,2,3-cd] pyrene	0.1	1	10	0.01	0.1	1
Group 2 Other PAH						
- naphthalene	0.1	5	50	0.2	2	20
- phenanthrene	0.1	5	50	0.2	2	20
- pyrene	0.1	10	100	0.2	2	20

* These values must be used in conjunction with the process described in Section 7.1.

** PAH designated as carcinogenic ("sufficient evidence") by IARC.

*** See explanation on page 36, paragraph in bold-faced text.

The groundwater values in Table 7.1 for the Group 1 carcinogenic PAH have been developed by the Working Group using the World Health Organization (WHO) Drinking Water Guideline for benzo[a]pyrene (B[a]P) as a reference value. The Value A numbers for these six PAH, have been selected as equal to the WHO B[a]P drinking water guideline. Value B numbers are intermediate values 10 times the Value A numbers. Value C numbers are 100 times the Value A numbers.

The A values for the Group 2 PAH, are based on routinely achievable detection limits. Values B and C are 10 and 100 times the value of A respectively. These A, B, C values for ground water are different than the values developed by the Quebec Ministry of the Environment.

Since no drinking water guidelines exist for any of the carcinogenic PAH except for B[a]P, the World Health Organization (WHO) guideline of .01 ug/L was adopted by the Working Group as being representative for the six carcinogenic PAH most commonly found in coal tar and as such was selected as the A value. However, since the WHO value was derived from a conservative, hypothetical, mathematical model that cannot be experimentally verified, the uncertainty surrounding this value is considerable and a variation of one tenth to ten times the value could exist.

For each of soil and ground water, Table 7.1 contains three threshold values, which determine levels of intervention. These are defined as:

Value A : This value represents the approximate achievable detection limits for PAH in soil.
In groundwater, value A is based on drinking water criteria as described in the bold-faced paragraph above.

Range A-B: The soil or groundwater is slightly contaminated. At this level of contamination, groundwater generally falls within the range of quality standards and criteria where they exist (see bold-faced paragraph above). However, it is worthwhile to investigate possible sources of

contamination, especially in the case of groundwater to ascertain whether new contaminants continue to enter the water. This may lead to intervention focusing on the soil, particularly if the water is used for drinking.

Usually at this level of soil or groundwater contamination, cleanup will not be necessary. However, should the land be redeveloped for especially sensitive purposes, e.g., residential or farming, it may be necessary to implement certain measures, such as the excavation of surficial layers of soil and/or the addition of a layer of clean soil.

Value B : This value is an intermediate value, approximately 5 to 10 times above value A.

Range B-C: The soil or groundwater is contaminated. At this level, contamination of groundwater clearly exceeds drinking water standards where they exist and can no longer be used for that purpose.

Although the soil is contaminated, it will not automatically be cleaned up, unless the effect of contaminants on the ground water necessitates such work.

However, restrictions on land use may be imposed when this level of contamination is observed in the soil. Restoration work may be necessary before the land is used for farming, residential or recreational purposes. Other less sensitive uses, e.g. industrial, commercial and so forth, may be contemplated without clean up being carried out. In all cases, the extent of the work required, e.g. the depth to which soil must be excavated and so on, will depend upon the nature of the contaminants, ultimate land use and the impact on groundwater and the environment in general.

Value C : This value is considered to be the level at which contamination is significant.

Range Above C : The soil or groundwater is contaminated. Groundwater cannot be used for drinking. Unless the water is decontaminated, it will have to be monitored closely.

Where the soil is contaminated, all uses of such land will be restricted. A thorough analysis must be conducted; in all likelihood, restoration will have to be undertaken before redevelopment occurs.

7.1.2 Recommended Application of Guidelines

The "ABC" approach provides a means to judge whether a site should be considered for further investigation and/or remediation. Depending on the use of the land, different investigative and/or remedial criteria will be considered.

Investigative criteria are values above which detailed investigation is required to assess the extent of contamination and nature of any hazards, to determine if any remedial actions should be undertaken.

Remedial criteria are values above which action is required to reduce exposure to humans or other biota. Action could include cleanup, other mitigation, and/or change in land use.

Residential or farming use

Investigative Criteria is Value A.

Remedial Criteria is Value B.

Commercial or industrial use

Investigative Criteria is the Value B.

Remedial Criteria is the Value C.

7.1.3 Disclaimer

Table 7.1 includes values for six (6) PAH which are designated as carcinogenic by IARC and values for three (3) other PAH which are usually present at high concentrations in coal tar.

The criteria defined for the concentration of PAH have been considered by the Working Group only for PAH related to coal tar sites and not to other sources. The application of these criteria to other PAH sources may or may not be appropriate.

7.2 AIR GUIDELINES

Few PAH air guidelines are available from other jurisdictions. While the provisional guidelines for B[a]P developed in Ontario were felt to have some application, these guidelines were judged not to be suitable for recommendation for adoption.

However, the Working Group felt that the basic approach developed by Ontario for air monitoring during cleanup at coal tar sites could be useful to CCREM agencies in site-specific cleanup.

The Ontario provisional air guidelines for B[a]P and the approach to air monitoring during coal tar cleanup are included as Appendix 3 for general information purposes.

It is currently not possible to predict site-specific background ambient PAH levels in air based on data collected elsewhere. Some of the reasons for this are: the volatility of PAH increases with temperature; and seasonal variability in other sources of PAH e.g. more residential heating in winter, and different traffic volumes.

7.3 SEDIMENT GUIDELINES

As with air guidelines, few PAH sediment guidelines have been developed elsewhere. The International Joint Commission have set a number in Great Lakes sediment for only B[a]P (Appendix 1).

Three case studies that demonstrate different approaches to establishing cleanup levels can be illustrated in actual clean-up situations of PAH contaminated sediments, two from Ontario, one from Nova Scotia.

At Chippawa Creek (near Hamilton, Ontario) sediments were contaminated with coal tar from an industrial site. Children swimming in adjacent areas of the creek developed dermatitis after contact with contaminated sediments.

A health based risk assessment to establish cleanup levels, using B[a]P as a surrogate for all the coal tar contaminants and focusing on the dermal exposure route, was unsuccessful. This lack of success was due to the absence of appropriate toxicity studies and exposure models which could extrapolate from dermal contact with contaminated sediments to evaluation of possible health risks. In lieu of the health based approach, background B[a]P concentrations in sediment were established for the cleanup site on the basis of a series of sediment samples collected on upstream transects and analysed for B[a]P. The sediments remaining at the site after cleanup contained B[a]P at levels similar to background.

A second approach to establishing cleanup levels is being considered at Port Stanley, Ontario (on Lake Erie), based on impact on aquatic invertebrates. The aquatic invertebrates are being considered representative of aquatic environmental effects in a tributary downstream of coal tar waste deposition. Similarly, in Sydney, Nova Scotia, effects on marine invertebrates are being considered as a basis for establishing cleanup levels. However, in Sydney, the concern is based on impact on human health and economic effects from the commercial lobster fishery.

Each of these approaches to setting cleanup criteria for sediments are very site-specific in nature, i.e., only B[a]P was considered at Chippawa Creek, total PAH at Port Stanley and Sydney; and the exposure routes are different, dermal exposure of children in Chippawa Creek, effects on aquatic life at Port Stanley and human exposure from contaminated lobster at Sydney.

The Working Group is of the opinion that the paucity of current information available and the very limited guidelines available for adoption, make it impossible to recommend interim sediment guidelines. The problem is further complicated by the fact that sites in Canada with affected sediments may be either marine or freshwater. It is not possible to extrapolate directly from one to the other because the physical/chemical properties of PAH, especially solubility which vary from fresh to salt water, as well as the PAH toxicity and the variety of exposure pathways.

Sediment guidelines should be established on a case-by-case basis considering the important exposure routes and most affected receptor.

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APPENDIX 1

COMPILATION OF AVAILABLE STANDARDS, GUIDELINES, OBJECTIVES, CRITERIA AND RECOMMENDATIONS FOR POLYCYCLIC AROMATICD HYDROCARBONS (PAH) IN AIR, WATER AND SOIL

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PREFACE

This compilation has been prepared as part of a project of the Toxic Substances Advisory Committee of the Canadian Council of Resource and Environment Ministers (CCREM) to derive interim guidelines for polycyclic aromatic hydrocarbons (PAH) in air, water, soil and sediments. This document presents in very cursory form, those guidelines, standards, criteria, etc. which were published or known, from a wide variety of sources including computerized data bases and personal communications. The information on the following pages is presented for comparison only. Care should be taken when reading the table on the following pages as no attempt has been made to standardize units. **It is recommended that the values listed not be used in any way without referring to original documentation or contacting officials of the agencies identified.**

Information on regulations, guidelines, etc. for PAH in the environment was gathered from the available literature, legislation, federal and provincial government departments. Information was sought on existing guidelines and regulations, and on guidelines and criteria which were being considered, developed or used as internal "rules of thumb" for management decision-making. Due to the very limited amount of published information regarding guidelines, etc. for PAH, a large proportion of the information gathered ws obtained through personal contacts, predominantly with members of the Ad Hoc Working Group on Interim PAH Guidelines.

Besides the use of computerized literature searches and personal contacts, a search was made of the IRPTC (International Registry of Potentially Toxic Chemicals) Legal Data Base, which includes records of regulations and guidelines for many European countries as well as Canada and the U.S.A. Access to this data base, which is maintained in Geneva, was available through the Environmental Health Directorate, Health & Welfare Canada.

The terms criterion, objective, guideline and standard have different meanings for different agencies. In this report, no

attempt has been made to standardize this terminology. Also, one agency may publish a variety of numerical values for the same medium, for different purposes and/or based on different rationales. The rationale for any given value could not be fully or adequately outlined in this document. Therefore, it is recommended that the values in this document be used for general comparison only and that source documents or sponsoring agencies be consulted before values are used for analytical or technical purposes.

MEDIUM	CHEMICAL	AGENCY	CONC.	UNITS	COMMENTS	REFERENCE
ir-occupational	Coal tar pitch volatiles	ACGIH	0.2	mg/m^3	Total of anthracene, BaP, phenanthrene, acridine, chrysene, pyrene	1
	Naphthalene	ACGIH	50	mg/m^3	TLV	1
		USSR	20	mg/m^3		15
		FRG, UK	50	mg/m^3	TLV	15
			75	mg/m^3	STEL	15
	B[a]P	OSHA	0.2	ug/m^3	Suggested in 1975	2
		USSR	0.150	ug/m^3	MAC	3
		Sweden	5.0	ug/m^3	TLV	3
			30.0	ug/m^3	STEL	3
	Phenanthrene	USSR	0.8	mg/m^3		15
	Pyrene	USSR	0.03	mg/m^3		15
r-ambient	B[a]P	USSR	1.0	ng/m^3	MAC 24 hr. av.	3
		Connecticut	0.1	ug/m^3	8 hr. av.	17
		Pennsylvania	0.7	ng/m^3	1 yr. av.	17
		Virginia	0.000	ug/m^3	24 hr. av.	17
		Ontario	0.3	ng/m^3	provisional guideline annual average for all sources	20
			0.22	ng/m^3	provisional guideline ann. av. for single source	20
			1.1	ng/m^3	24 hr. average provisional ambient air quality criteria (AAQC)	20
			3.3	ng/m^3	0.5 hr. provisional single source point of impingement guideline	20

MEDIUM	CHEMICAL	AGENCY	CONC.	UNITS	COMMENTS	REFERENCE
Air-ambient continued	Naphthalene	USSR	0.003	mg/m^3	24 hour av.	15
		Connecticut	1.0	mg/m^3	8 hour av.	17
		Maryland	14.0	ug/m^3	24 hour av.	17
		Nevada	1.19	mg/m^3	8 hour av.	17
		New York	166.7	ug/m^3	1 year av.	17
		Virginia	800.0	ug/m^3	24 hour av.	17
		California	0.0018	mg/m^3	Applied Action Level for human receptor 24 hour av.	16
		Ontario	22.5	ug/m^3	24 hour av. provisional ambient air quality criteria (AAQC)	20
			36	ug/m^3	0.5 hr. provisional point-of-impingement guideline	20
	Fluorene	Connecticut	50.0	ug/m^3	8 hour av.	17
		California	0.0019	mg/m^3	Applied Action Level for human receptor 24 hour av.	16
	Pyrene	California	0.0019	mg/m^3	Applied Action Level for human receptor 24 hour av.	16
	Phenanthrene	California	0.0019	mg/m^3	Applied Action Level for human receptor 24 hour av.	16
	Acenaphthylene	California	0.0019	mg/m^3	Applied Action Level for human receptor 24 hour av.	16
	Acenaphthene	California	0.0019	mg/m^3	Applied Action Level	16

MEDIUM	CHEMICAL	AGENCY	CONC.	UNITS	COMMENTS	REFERENCE
-ambient tinued	Anthracene	California	0.0019	mg/m^3	Applied Action Level for human receptor 24 hour av.	16
er-ambient	Total carcinogenic PAH	USEPA	2.8	ng/l	With fish and water consumption	4
			3.5	ng/l	With water consumption only	4
			31.1	ng/l	With fish consumption only	4
	Total PAH		300	ug/l	LOEL acute-saltwater aquatic life	4
	Acenaphthene	USEPA	1700	ug/l	LOEL acute-freshwater aquatic life	5
			520	ug/l	LOEL chronic-freshwater aquatic life	5
			970	ug/l	LOEL chronic-saltwater aquatic life	5
			20.0	ug/l	To conotrol undesirable tate and odour	5
		California	0.019	mg/l	Applied Action Level for human receptor	16
	Acenaphthylene	California	0.019	mg/l	Applied Action Level for human receptor	16
	Anthracene	California	0.019	mg/l	Applied Action Level for human receptor	16
	Fluoranthene	California	0.019	mg/l	Applied Action Level for human receptor	16

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MEDIUM	CHEMICAL	AGENCY	CONC.	UNITS	COMMENTS	REFERENCE
Water-ambient continued	Fluoranthene	USEPA	42.0	ug/l	Human health-with fish consumption	6
			200	ug/l	Human health-without fish consumption	
			54	ug/l	Human health-consumption of fish only	6
			3980	ug/l	LOEL acute-freshwater aquatic life	6
			40	ug/l	LOEL acute-saltwater aquatic life	6
			16	ug/l	LOEL chronic-saltwater aquatic life	6
	Fluorene	California	0.019	mg/l	Applied Action Level for human receptor	16
	Naphthalene	USEPA	2300	ug/l	LOEL acute-freshwater aquatic life	7
			620	ug/l	LOEL chronic-freshwater aquatic life	7
			2350	ug/l	LOEL acute-saltwater aquatic life	7
		USSR	0.01	mg/l	surface water	15
			0.004	mg/l	surface water for fishing	15
		California	0.018	mg/l	Applied Action Level for human receptor	16
	Pyrene	California	0.019	mg/l	Applied Action Level for human receptor	16
	Phenanthrene	California	0.019	mg/l	Applied Action Level for human receptor	16

MEDIUM	CHEMICAL	AGENCY	CONC.	UNITS	COMMENTS	REFERENCE
ater-ambient ontinued	B[a]P	USSR	0.005	ug/l	Regulated MAC in surface water	3
		GLSAB	0.01	ug/l		
			1.0	ug/l	Surface water to receive intensive treatment before drinking	15
		WHO	0.01	ug/l	Guideline	9
	Total PAH	EEC	0.2	ug/l	MAC	18
	Total of fluoranthene, benzo[k] fluoranthene, B[a]P Benzo[ghi]perylene, indeno[1,2,3-cd] pyrene, pyrene	WHO	0.2	ug/l	Guideline	9